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Westinghouse
Hanford Company Richland, Washington

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R. C. Knight
D. L. Greenslade

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**Westinghouse
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P.O. Box 1970
Richland, Washington 99352

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IRRADIATION TESTING OF A NIOBIUM-MOLYBDENUM DEVELOPMENTAL THERMOCOUPLE

R. Craig Knight
Westinghouse Hanford Company
P. O. Box 1970
Richland, Washington 99352
(509) 376-5419

David L. Greenslade
Westinghouse Hanford Company
P. O. Box 1970
Richland, Washington 99352
(509) 376-5601

Abstract

A need exists for a radiation-resistant thermocouple capable of monitoring temperatures in excess of the limits of the chromel/alumel system. Tungsten/rhenium and platinum/rhodium thermocouples have sufficient temperature capability but have proven to be unstable because of irradiation-induced decalibration. The niobium/molybdenum system is believed to hold great potential for nuclear applications at temperatures up to 2000 K. However, the fragility of pure niobium and fabrication problems with niobium/molybdenum alloys have limited development of this system. Utilizing the Fast Flux Test Facility, a developmental thermocouple with a thermoelement pair consisting of a pure molybdenum and a niobium-1%zirconium alloy wire was irradiated for 7200 hours at a temperature of 1070 K. The thermocouple performed flawlessly for the duration of the experiment and exhibited stability comparable to a companion chromel/alumel unit. A second thermocouple, operating at 1375 K, is currently being employed to monitor a fusion materials experiment in the Fast Flux Test Facility. This experiment, also scheduled for 7200 hours, will serve to further evaluate the potential of the niobium-1%zirconium/molybdenum thermoelement system.

INTRODUCTION

Accurate temperature measurement of nuclear reactor experiments requires specific capabilities. Thermocouples must be immune to irradiation damage that would alter the electro-chemical properties of the thermoelement wire pair. For low-to-moderate temperature applications, chromel/alumel (Type K) thermocouples have proven to be extremely reliable. However for extended service at temperatures in excess of approximately 1350 K, other systems must be considered.

Tungsten/rhenium (W/Re) and platinum/rhodium (Pt/Rh) thermocouples have higher temperature capability. However, these systems are subject to significant decalibration caused by irradiation effects including transmutation of the base materials (Browning and Miller 1962, and Heckelman and Kozar 1972) and second phase precipitation (Williams et al. 1983). Figure 1 shows the negative drift that occurred with a W/Re thermocouple during an irradiation experiment in the Fast Flux Test Facility (FFTF), a 400-MW_t liquid metal cooled reactor located at the Hanford Site near Richland, Washington. This particular experiment operated at a temperature of 1350 K (Greenslade et al. 1990).

FIGURE 1. Thermocouple Performance in the FFTF Materials Open Test Assembly Canister 5C. Thermocouples were Exposed to a Fast Fluence ($E > 0.1$ MeV) of 4.0×10^{22} n/cm². Note: Type C is an Unofficial Designation for a W5%Re/W26%Re Thermocouple.

Niobium and molybdenum (Nb-Mo) alloys have previously been considered for thermocouple applications because these refractory metals have low neutron capture cross sections as well as high temperature capability. Significant work on Nb/Mo thermocouples occurred in France (Schley and Metauer 1982). Although the system showed significant potential, problems with alloy fabrication hampered this effort. Further work at the Idaho National Engineering Laboratory (INEL) also encountered fabricability problems (Wilkins 1988).

One reason that previous efforts focused on Nb/Mo alloys rather than pure metals was because the alloys exhibit better electromotive force (EMF) potential at temperatures above approximately 1750 K (Schley and Metauer 1982). However, 1750 K is considerably higher than the range of Type K thermocouples and encompasses the requirements for many nuclear experiments of interest. Because of this fact, as well as the availability of materials, the initial work at Westinghouse Hanford Company concentrated on the pure metals. However, preliminary furnace tests with thermocouple probes utilizing 0.25-mm-diameter thermoelement wires at temperatures up to 1675 K resulted in early failure attributed to excessive grain growth in the pure niobium wire.

As shown in Figure 2, the wire was transformed into a string of single crystals subject to transgranular failure caused by thermal stresses during temperature transients. The molybdenum wire did not exhibit such rapid grain growth, and no failures were experienced in this component.

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FIGURE 2. Longitudinal Photomicrograph of 0.25-mm-diameter Niobium Wire from Test Thermocouple, Furnace Aged for 260 Hours at 1475 K with a Brief Excursion to 1675 K (250X).

These results led to investigation of the relatively common Nb alloy, niobium-1% zirconium (Nb-1%Zr) used in conjunction with pure molybdenum to form the thermoelement wire pair.

EXPERIMENT DESIGN

Having limited resources, we decided to utilize materials on hand to devise a thermocouple experiment for testing in the FFTF. Both Nb-1%Zr and pure Mo wire of 0.25-mm diameter were available in our laboratory inventory. High purity (99.8%) alumina insulators, sized to fit the 0.25-mm wires, were also available. Unalloyed Mo sheath tubing, 1.8-mm OD x 1.2-mm ID, was fabricated from metal powder by a commercial supplier.

During testing of the pure metal system, a calibration test was run to a temperature of 1950 K utilizing a Type B (Pt/Rh) thermocouple for comparison. The EMF curve generated agreed closely with previous data (Wilkins 1988).

Upon incorporation of the Nb-1%Zr wire, we decided to perform a more precise calibration experiment. A thermocouple probe consisting of Nb-1%Zr/Mo wires, alumina insulators, and Mo sheath approximately 0.3 meters in length was assembled and inserted into a high temperature vacuum furnace. Pure copper lead wires were soldered to the thermoelement wires and run through an ice bath to a calibrated voltmeter. Millivolt readings were taken while the furnace temperature was cycled to 1450 K and back. The EMF curve shown in Figure 3 resulted. The curve was found to vary only slightly from that for the pure metals.

FIGURE 3. Voltage Versus Temperature Curve for Alumina Insulated, Molybdenum Sheathed, Niobium-1%Zirconium/Molybdenum Thermocouple.

To obtain meaningful data in an irradiation experiment, it was necessary to find a way to generate a temperature gradient over the length of the Nb/Mo thermoelement wires. Since the system was experimental, we ruled out the use of a thermocouple to monitor an actual FFTF experiment. However, a method was devised to take advantage of nuclear heating of the molybdenum thermocouple sheath by enclosing the thermocouple probe in a vacuum chamber. The low thermal conductivity in vacuum would allow the probe to operate well above the temperature of the surrounding coolant. As shown in Figure 4, computer analysis disclosed that this method could be expected to generate a temperature of up to 1150 K, depending on the length of the thermocouple probe. Using this technique, all that remained was to find a vehicle to carry the test thermocouple into the core of the FFTF. A Materials Open Test Assembly (MOTA) (Greenslade et al. 1986) was selected.

FIGURE 4. Computer Analysis Depicting Anticipated Performance of a Nb-1%Zr/Mo Test Thermocouple in the FFTF at Core Mid-Plane, Row 4 Position. Mo-Sheathed Probe is Enclosed in Stainless Steel Vacuum Containment.

FABRICATION AND INSTALLATION

Figure 5 shows the construction of the test thermocouples. To achieve the maximum operating temperature (see Figure 4), a probe length of 7 cm was selected. As noted in Figure 5, the Nb-1%Zr/Mo thermocouple wires were spliced to Type K wires adjacent to, but outside of, the vacuum containment. In this way, the dissimilar metal splices could be maintained at a constant (coolant) temperature--an important criteria for thermocouple accuracy. Standard MOTA Type K thermocouple cables were used. A vacuum of approximately 5×10^{-5} torr was achieved in the stainless steel enclosure by performing the final end cap weld in an electron beam welder. The thermocouple probe was backfilled at room temperature with one atmosphere of high purity helium.

FIGURE 5. Schematic Showing the Component Parts of the Test Thermocouples.

Two test thermocouples and one identically constructed Type K thermocouple were fabricated. The purpose of the Type K thermocouple was to provide comparative data (computer analyses showed that replacement of the Nb/Mo wires with Type K wires would have negligible effect on the temperature achieved by nuclear heating).

Due to space limitations, there was room for only two special thermocouples on the selected MOTA experiment. One Nb-1%Zr/Mo and one Type K thermocouple were affixed to the MOTA test train at the FFTF core mid-plane location. In addition, a standard MOTA Type K thermocouple was placed near the location of the dissimilar metal splices on the test thermocouple to accurately monitor the reactor coolant temperature at that point.

The corrected millivolt reading of the Nb-1%Zr/Mo test thermocouple assembly, including the Type K extension cable, was obtained using the following equation:

$$MV_c = MV_t - MV_{ks} + MV_{ns}$$

where MV_c is the corrected voltage, MV_t is the total indicated voltage, MV_{ks} is the voltage indicated by the Type K thermocouple at the splice, and MV_{ns} is the corresponding voltage that a Nb/Mo thermocouple located at the splice would indicate (see Figure 3).

During operation, the MOTA data acquisition system automatically performed the above calculation and converted the voltage outputs directly to temperature. Data points were taken every 5 minutes and logged once per hour for the duration of the test. The experiment began operation in the FFTF in January 1990 and concluded in March 1991, with 300 full power days (FPD) achieved.

RESULTS AND DISCUSSION

Both test thermocouples performed without failure for the duration of the MOTA irradiation. As shown in Figure 6, there was an initial period of instability before the two thermocouples began to indicate similar steady-state temperatures. The final temperature achieved averaged 1070 K, approximately 100 degrees less than predicted in Figure 4. We also observed that the Type K thermocouple indicated temperature continued to fluctuate by

several degrees throughout the experiment, while the Nb-1%Zr/Mo test thermocouple was much more stable.

FIGURE 6. Performance of the Test Thermocouples in the FFTF During 300 Full Power Days Operating Cycle. Thermocouples were Exposed to a Fast Fluence of 7.5×10^{22} n/cm².

As noted in Figure 6, the average temperature of both thermocouples continued to decline slightly over the duration of the test. Over the final 100 FPD, the average temperature of the Nb-1%Zr/Mo thermocouple decreased from 1075 K to 1067 K--a total of 8 degrees. During this same period, the average temperature of the Type K thermocouple decreased by a similar amount, from 1070 K to 1061 K--a total decline of 9 degrees.

The causes of the instability at the start of irradiation, the continued oscillation of the Type K thermocouple, and the overall steady decrease in operating temperature are believed to include one or more of the following:

- A less-than-optimum vacuum atmosphere within the stainless steel outer enclosure;

- Radiation-caused changes in the emissivity of the component surfaces (OD of the Mo sheath and ID of the stainless steel vacuum containment tube); and
- Movement of the thermocouple probe within the vacuum containment tube caused by thermal stresses.

The degree of vacuum achieved in the containment tube by making the final end cap weld in an electron beam welder is questionable. Because of the tight fit-up of components, it is possible that some amount of air may have remained in the containment tube after welding. This would lead to conduction losses, oxidation, and surface emissivity changes during operation.

The temperature oscillations of the Type K thermocouple could be the result of movement of the probe within the vacuum containment. Although radiographic inspection showed excellent as-fabricated alignment, thermal stresses may have caused movement of the probe close to, or in intermittent contact with, the tube ID, resulting in substantial temperature variations.

CURRENT PROGRAMS

Because of the success of this experiment, we decided to use a Nb-1%Zr/Mo thermocouple to monitor a high-temperature experiment scheduled for irradiation in a follow-on FFTF MOTA.

The experiment, designed to evaluate candidate fusion reactor materials, required thermocouples capable of monitoring temperatures of up to 1475 K. Because of the potential to exceed the safe operating limit of Type K thermocouples, agreement was reached to place a Nb-1%Zr/Mo thermocouple with a Type K thermocouple in the MOTA test canister.

For this experiment, the Nb/Mo thermocouple consisted of the following materials:

- Thermoelement - Nb-1%Zr and unalloyed Mo, 0.25-mm diameter;
- Insulators - 99.8% purity polycrystalline alumina;
- Sheath - W22%Re, 1.6-mm OD x 1.2-mm ID; and
- Lead cable - 2.4-mm OD stainless steel sheathed Type K.

The Type K thermocouple was identically fabricated except for the thermoelement wires which were 0.25-mm OD chromel/alumel.

This experiment commenced operation in May 1991. Figure 7 shows the temperature history for the first operating cycle which totaled 53 FPDs. As shown, the maximum experimental temperature achieved was approximately 1375 K. Both the Nb-1%Zr/Mo and Type K thermocouples operated flawlessly, with the latter averaging 42 K less than the experimental thermocouple because of the location of the thermocouples within the MOTA canister.

FIGURE 7. Comparison of the Performance to Date of a Test Nb-1%Zr/Mo Thermocouple with a Type K Thermocouple in a Fusion Materials Experiment in the FFTF.

Figure 7 shows a perceptible decline in the temperature indicated by both thermocouples. This is because of predicted burnup of the experimental materials and is not an indication of thermocouple decalibration. In fact, the actual difference between the two thermocouples varied only 3 degrees throughout the operating cycle--further proof that the Nb-1%Zr/Mo system is practically immune to radiation-induced decalibration.

The fusion materials experiment is scheduled to continue for a total irradiation cycle of 300 FPD.

CONCLUSIONS

The passive experiment demonstrated the stability of the Nb/Mo thermocouple system under severe irradiation conditions at an intermediate temperature. At core mid-plane in the FFTF, the thermoelement wires were subjected to a total fluence of 12.5×10^{22} n/cm² and a fast fluence of 7.5×10^{22} n/cm². The slight decrease in temperature noted as the irradiation progressed was concluded to be unrelated to nuclear transmutation or other irradiation effects as verified by the similar behavior of the Type K thermocouple.

Type K thermocouples have performed flawlessly in the FFTF under similar conditions of temperature and fluence in numerous irradiation tests.

The results also demonstrated the practicality of the experimental method. Although some refinement of the fabrication techniques is needed, the theory proved to be sound and the manufacturing cost was minimal. In addition, there was no risk to the operation of the plant or to other reactor experiments. While there are no current plans, the method could readily be used in the future to evaluate other developmental thermocouple systems.

The follow-on experiment will provide additional data regarding the viability of the Nb/Mo thermocouple system.

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